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Modelling of particulate matter concentrations and source contributions in the Helsinki Metropolitan Area in 2008 and 2010

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We refined an urban-scale dispersion modelling system by adding a road dust suspension model, FORE. The deterministic modelling includes both vehicular exhaust emissions (including cold start and cold driving) and suspended road dust. The urban scale modelling system was used in combination with the regional scale chemical transport model LOTOS-EUROS, for 2008, and the measured regional background concentrations, for 2010. The predictions were compared against measured concentrations of $PM_{2.5}$ and PM_{10}. $PM_{2.5}$ concentrations were slightly and the PM_{10} concentrations substantially under-predicted in 2008, mainly due to the under-predicted regional background concentration. Source contributions of suspended road dust varied from 2% to 8% and from 12% to 38% for $PM_{2.5}$ and PM_{10}, respectively. Long-range transported contributions at the urban traffic stations were 72% to 92% for $PM_{2.5}$ and 50% to 83% for PM_{10}.

Introduction

There is substantial evidence for short-term and long-term effects of ambient air particulate matter, especially $PM_{2.5}$, on cardiovascular and respiratory systems. There is also emerging evidence for ambient air particulate matter causing premature births, affecting lung-function development in children, and promoting atherosclerosis and cognitive impairment (Anonymous 2013) as well as autism (Volk *et al*. 2013). Particulate matter (PM) levels that have been estimated to be relatively safe, using guideline values by agencies, such as WHO, can contribute to higher

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rates of cognitive decline, stroke, and heart attack (Devi 2012). In addition, the current EU limit value of 25 μg m^{-3} for the annual average concentrations of $PM_{2.5}$ is substantially higher than the WHO Air Quality Guideline of 10 μg m^{-3}.

Particulate matter and its source contributions in the Nordic countries have recently been studied, for example, by Gidhagen *et al*. (2013). They modelled residential outdoor particulate matter (PM_1 and PM_{10}) levels in the whole of Sweden, including long-range transport, local traffic exhaust and road dust in 2004 and 2005 using the SIMAIR modelling system. They found that long-range transport commonly in dominated the average Swedish residential PM_1 and PM_{10} levels; however, the contributions from urban and local traffic sources could dominate in case of residences that were close to very busy roads.

Particulate matter source contributions were also studied in other European cities. For example, Singh *et al*. (2014) presented an analysis of $PM_{2.5}$ in 2008 in London, comparing observed concentrations with predictions of the OSCAR Air Quality Assessment System. They produced estimates for the urban traffic increments, as well as for the total urban increments of $PM_{2.5}$ in London in 2008, for several types of environments. They concluded that both the urban increment and the traffic contribution to total $PM_{2.5}$ were significant, although spatially heterogeneous across London. They estimated that in London approximately two thirds of the traffic increment originated from exhaust emissions and most of the rest was from brake and tyre wear. Keuken *et al*. (2013) studied $PM_{2.5}$ and PM_{10} and their chemical composition in Rotterdam in 2010 and 2011. They estimated that urban background of $PM_{2.5}$ and PM_{10} was dominated by the regional background, and the primary and secondary $PM_{2.5}$ emissions by urban sources contributed less than 15% to the ground-level concentrations in Rotterdam.

The emissions originated from road dust, suspended by road traffic were modelled by Kauhaniemi *et al*. (2011) with the model FORE (Forecasting Of Road dust Emissions). The model was evaluated in combination with the street canyon model OSPM for PM_{10} in a street canyon environment in Helsinki. The model combination was found to reproduce fairly well the seasonal variation in the PM_{10} concentrations during a measurement campaign of four months in winter and spring, also in the presence of an extended use of studded tyres and anti-skid street treatments. Kauhaniemi *et al*. (2014) then compared the PM_{10} predictions of the FORE model with another road suspension emission model, the NORTRIP (NOn-exhaust Road TRaffic Induced Particle emissions) against the road suspension measurements onboard a mobile van, in two urban area street segments in Helsinki. Both models were found to substantially underpredict emissions.

In this study, we included the road dust suspension model FORE in an urban scale modelling system, to be used in a more extensive urban area. The urban modelling system was used in combination with the chemical transport model LOTOS-EUROS (Schaap *et al*. 2008). The modelled results were compared against the $PM_{2.5}$ and PM_{10} measurements from two sources, HSY (Helsinki Region Environmental Services Authority) air quality measurement network and the ESCAPE (European Study of Cohorts for Air Pollution Effects) campaign in the Helsinki Metropolitan Area. We used the results of two years, 2008 and 2010. The aims of this study were (i) to compare the predictions of the combined urban and regional modelling system against available measured data for both $PM_{2.5}$ and PM_{10}, and (ii) to evaluate the contributions to the predicted concentrations of vehicular traffic, explicitly allowing for both exhaust and road suspension emissions, and regional and long-range transport.

Material and methods

The Helsinki Metropolitan Area (HMA) comprises four cities; Helsinki, Espoo, Vantaa and Kauniainen. The total population in the HMA is approximately 1.1 million, while the population of Helsinki is about 0.6 million inhabitants. The most important local source of the PM mass fractions is vehicular traffic, with smaller contributions from shipping and harbour operations, industrial sources, small-scale combustion (Soares *et al*. 2014), and aviation.

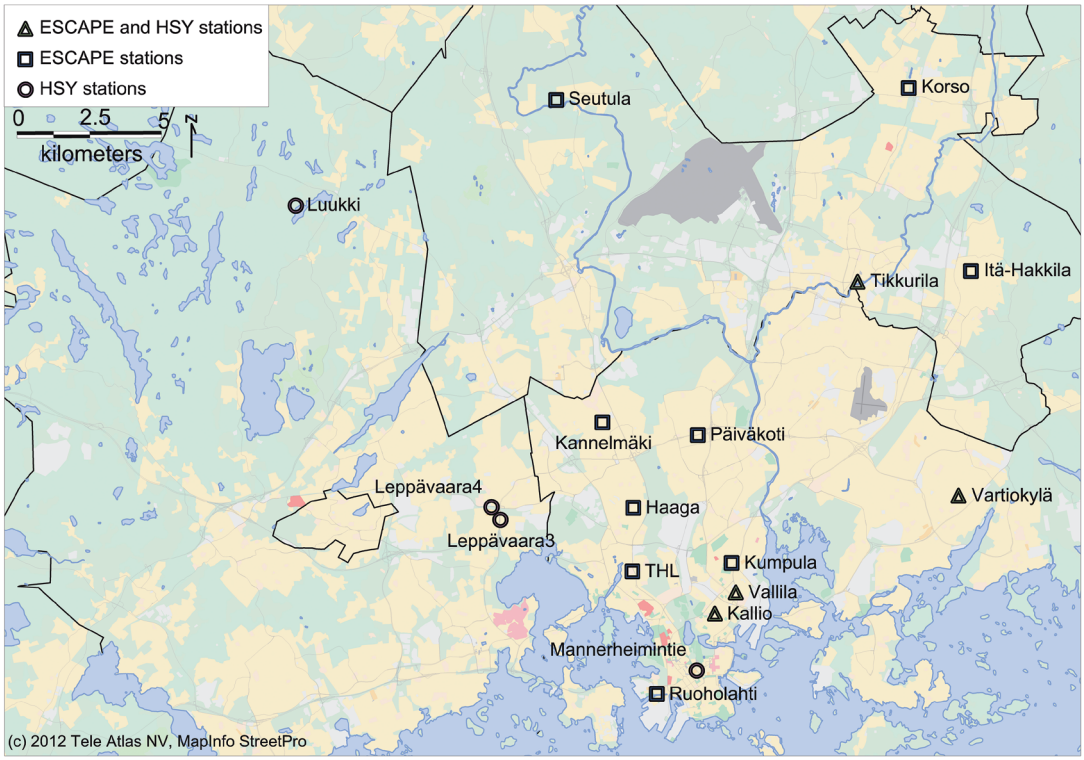

Fig. 1. Locations of the measurement sites in the Helsinki Metropolitan Area (HMA) in 2008 and 2010. Some sites are joint HSY (Helsinki Region Environmental Services Authority) and ESCAPE (European Study of Cohorts for Air Pollution Effects) sites, and some are ESCAPE or HSY sites only. The city borders are marked with black lines. The built-up areas are in yellow and red, and green areas are in green.

Experimental data

Meteorological data

We used the synoptic weather observations from the stations at the Helsinki-Vantaa airport (18 km north of the Helsinki city centre) and Kumpula (5 km north of the city centre), and sounding observations from Jokioinen (90 km northwest of Helsinki) for the years 2008 and 2010. These observations were required as input for a meteorological pre-processing model.

Concentration data

We used the hourly time series of $PM_{2.5}$ and PM_{10} measured by the HSY air quality measurement network (Fig. 1), including both PM fractions for the year 2008 and $PM_{2.5}$ for 2010. The measurement methods used in our study (Table 1) were calibrated to ensure that the concentrations measured using different instruments are inter-comparable. The calibration functions for the continuous measurement methods for $PM_{2.5}$ against a reference method were determined for Helsinki during 2007–2008 (Waldén *et al*. 2010). The hourly observation data were pre-processed by removing negative concentration values from the analysis. We addressed only the $PM_{2.5}$ observations for the year 2010, as our main focus was on the most health-affecting pollutants.

In addition, we used the $PM_{2.5}$ measurement data for the HMA from the ESCAPE project (Eeftens *et al*. 2012) collected in 2010. The ESCAPE data set contained two weekly concentration averages. Between February 2010 and January 2011, there were 24 such measurement periods for the urban background site in Kallio, and for the other sites from two to three weekly measurement periods. All ESCAPE particulate matter samples were collected using Harvard impactors, designed to

collect $PM_{2.5}$ (Hoek *et al*. 2002). The ESCAPE site types were urban street, urban background and regional background. The ESCAPE observations in HMA in 2010 were made only for $PM_{2.5}$ (instead of both PM mass fractions).

The PM_{10} concentrations measured at the regional background station of Virolahti, located approx. 150 km E-NE of Helsinki, were used in a comparison of observed and predicted long-range-transported background for the year 2008. The Virolahti PM_{10} time series was measured with Eberline FH 62 IR, and uncalibrated data were used.

In the evaluation of the regionally- and long-range-transported $PM_{2.5}$ background concentrations (LRT) for 2010, we used the observed concentrations at the regional background station of Luukki.

Mathematical models and methods

Meteorological pre-processing

Measured meteorological data were analysed using the meteorological pre-processing model MPP-FMI that has been adapted for an urban environment (Karppinen *et al*. 2000a). The computation was based on a combination of the data from the stations at the Helsinki–Vantaa airport, Helsinki-Kumpula (3-hour synoptic weather observations) and Jokioinen (soundings) for the years 2008 and 2010. The MPP-FMI model is based on the energy budget method of van Ulden and Holtslag (1985). The model utilises meteorological synoptic and sounding observations, and its output consists of the hourly time series of the relevant atmospheric turbulence parameters and the boundary layer height.

The output of the MPP-FMI model contains the meteorological data that are needed for the dispersion modelling, including temperature, wind speed, wind direction, Monin-Obukhov length, friction velocity, and boundary layer height. The road suspension emission model utilizes hourly-averaged pre-processed meteorological data for the following parameters: temperature, dew-point temperature, precipitation, relative humidity, wind speed, global radiation and total cloud cover.

Table 1. The measurement sites of HSY used in this study, their site classifications and measurement methods of $PM_{2.5}$ and PM_{10} in 2008 and 2010. Abbreviations of the site classifications: Reg = regional, bg = background, tr = traffic, U = urban and Res = residential.

Year	Pollutant	HSY site	Site classification	Measurement devices*
2008	$PM_{2.5}$	Luukki	Reg bg	TEOM 1400a + FDMS, TEOM 1400A from 17 Jan. 2008
		Kallio	U bg	TEOM 1400 AB, FH 62 I-R from 10 Nov. 2008
		Mannerheimintie	U tr	FH 62 I-R
	PM_{10}	Virolahti	Reg bg	FH 62 I-R
		Kallio	U bg	TEOM 1400 AB
		Leppävaara3	U tr	TEOM 1400 AB until 19 Dec. 2008, then in 2008 Grimm 180
		Mannerheimintie	U tr	FH 62 I-R
		Tikkurila3	U tr	Grimm 180
		Vallila1	U tr	FH 62 I-R
2010	$PM_{2.5}$	Luukki	Reg bg	FH 62 I-R
		Kallio	U bg	TEOM 1400 AB
		Vartiokylä	Res bg	Grimm 180
		Tikkurila3	U tr	Grimm 180
		Leppävaara4	U tr	Grimm 180
		Mannerheimintie	U tr	FH 62 I-R

* Calibration: PM_{10} calibration functions reported by the manufacturers of the measurement devices (Sillanpää *et al.* 2002, LUBW 2005). $PM_{2.5}$ concentration: β-radiation absorption: (Eberline FH62-IR $\times$ 1.35) – 0.73 μg m^{-3}, tapered element oscillating microbalance: (TEOM $\times$ 1.25) + 1.56 μg m^{-3}, laser scattering: (Grimm $\times$ 0.75) – 0.31 μg m^{-3}. PM_{10} concentration: Grimm $\times$ 0.82.

Evaluation of urban emissions in the Helsinki Metropolitan Area

Evaluation of traffic flows and vehicular exhaust emissions

The emission inventory included exhaust emissions from vehicular traffic for the network of roads and streets in the HMA. In this study, approximately 4300 road and street links were included in the computations. The traffic volumes and average travel speeds of each traffic link were calculated using the EMME/2 transportation planning system (INRO, 1994) for the year 2005. The emission factors of cars were based on national measurements.

The emission inventory consisted of average hourly emissions for each line source during a year, separately for weekdays, Saturdays and Sundays. Cold start and cold driving emissions were also taken into account, and they were modelled with coefficients based on laboratory emission measurements (Kauhaniemi *et al*. 2008). These coefficients were estimated separately for weekdays and weekends, and they also took into account the temperature of the ambient air and the pre-heating of vehicle engines (Kauhaniemi *et al*. 2008).

The emission inventory for 2005 was applied for 2008 and 2010, by scaling the emission in each road link in 2005. The scaling was done by multiplying the emissions in 2005 by the ratio of the total exhaust emission values in the HMA in 2008 or 2010 to that in 2005. The total exhaust emissions were extracted from the national traffic exhaust emissions and energy consumption archive (lipasto.vtt.fi).

Evaluation of vehicular suspension emissions

We used the FORE model 'Forecasting of Road Suspension Emissions' (Kauhaniemi *et al*. 2011). This model is based on the particulate suspension emission model by Omstedt *et al*. (2005); that model was revised, to be applicable also to operational forecasting of air quality. The FORE model was evaluated against experimental data and another road suspension model by Kauhaniemi *et al*. (2011 and 2014). The suspension emission (μg m^{-1} s^{-1}) of a line source is defined as the product of the number of vehicles per hour and the suspension emission factor (μg vehicle^{-1} m^{-1}). The emission factors for suspension are modelled by considering the moisture content of the road surface and the particles originating from the wear of pavement and from traction sand.

The emission factor for the suspension of road dust is a product of the so-called reference emission factors, the reduction factor for the moisture content, and a weighted sum of the contributions originated from particles from the wear of pavement and from the traction sand (Kauhaniemi *et al*. 2011). The emissions of brake, tyre and clutch wear were not taken into account in the present model. Kupiainen *et al*. (2015) estimated the emissions of traffic related non-exhaust $PM_{2.5}$ and PM_{10} for HMA for the period 2008–2012. They estimated road dust suspension with a calculation method based on the results and experiences of the NORTRIP (Denby *et al*. 2013) and REDUST (www.redust.fi) projects. Kupiainen *et al*. (2015) estimated from the literature that PM_{10} and $PM_{2.5}$ emissions originating from tyre, brake and pavement wear were roughly 0.5 and 0.7 times, respectively, of those estimated for road-dust suspension.

Evaluation of the influence of other local emission categories

The evaluation of local urban emissions in this study included vehicular exhaust emissions and suspension emissions. Soares *et al*. (2014) and Saarnio *et al*. (2012) evaluated the contributions of other local sources to the concentrations of $PM_{2.5}$ in the HMA.

The contributions of shipping and major stationary-source emissions to the total emissions of $PM_{2.5}$ in the HMA in 2008 were approximately 8% and 28%, respectively (Niemi *et al*. 2009).

In the HMA, residential buildings and household water are heated by electricity (33%, energy consumption by household appliances), district heating (29%), and small-scale combustion of mainly wood (23%). The contribution of small-scale combustion to the total $PM_{2.5}$ emissions in the HMA in 2009 was estimated to be 23%

(Niemi *et al*. 2009). However, it was not possible to include the effects of small-scale combustion in this study, as the spatial distribution of the emission data was not known with sufficient accuracy.

The emissions of $PM_{2.5}$ originating from aviation in the HMA were about 0.1% of the total $PM_{2.5}$ emissions in the area in 2008 (Niemi *et al*. 2009). They can therefore be considered negligible.

Urban scale dispersion modelling

The urban-scale dispersion of vehicular emissions was evaluated with the CAR-FMI model (Contaminants in the Air from a Road, Finnish Meteorological Institute) (Härkönen 2002, Karppinen *et al*. 2000c, Kukkonen *et al*. 2001a, 2001b). The model is a Gaussian finite line source model, in which the dispersion parameters are modelled as a function of Monin-Obukhov length, friction velocity, and boundary layer height. The concentration values were computed for the HMA. Street canyon dispersion modelling was not performed in this study. No chemical reactions or aerosol transformation processes were included in the calculations. The receptor grid intervals ranged from 20 m in the vicinity of the major roads to 500 m on the outskirts of the area. The concentration values were computed at more than 18 and 24 thousand receptor points for 2008 and 2010, respectively.

The modelling system containing the CAR-FMI model was evaluated against the measured data of urban measurement networks for gaseous pollutants (e.g., Karppinen *et al*. 2000b and Kousa *et al*. 2001) and for $PM_{2.5}$ (Kauhaniemi *et al*. 2008, Sokhi *et al*. 2008, Singh *et al*. 2013) in the HMA and London. The performance of the CAR-FMI model was also evaluated for gaseous pollutants against the results of a field measurement campaign and other roadside dispersion models (Kukkonen *et al*. 2001a, 2001b, Oettl *et al*. 2001, Levitin *et al*. 2005).

Regional scale modelling

For regional-scale modelling, a new emission inventory compiled in the TRANSPHORM project was used for the EU-wide transport activities, supplemented by non-transport activities. The baseline emission data contain the following substances: NO_x, SO_2, non-methane volatile organic compounds (NMVOC), CH_4, NH_3, CO, PM_{10}, $PM_{2.5}$, EC (elemental carbon), B[*a*]P (benzo[*a*]pyrene) and particulate number. The inventory includes particles in the 10–300 nm size range. The particle number emission inventory includes only anthropogenic sources; emissions from natural sources such as wildland fires, windblown dust and sea salt are not included. The inventory also does not include vegetation-related emissions, or the formation of PNCs from biogenic VOCs.

The regional-background PM concentrations for 2008 were evaluated using the LOTOS-EUROS model (Schaap *et al*. 2008). We used the predictions of this particular chemical transport model, as the computations of this study are part of a more extensive evaluation in the target cities within the TRANSPHORM project. Also, the data measured at the HMA regional background monitoring station of Luukki did not include the measurements of PM_{10}. Therefore, as the regional background concentration values for 2008 we used the LOTOS-EUROS-predicted hourly $PM_{2.5}$ and PM_{10} concentration values for the 7×7 km^2 grid square, corresponding to the regional background station at Luukki.

The long-range transported contribution for 2010 was estimated using the measured hourly concentration values at the regional background station of Luukki. We selected this option, as (i) we expected this procedure to be the most accurate method for estimating the regional background contribution, and (ii) this choice facilitated a comparison of the accuracy of the two methods for evaluating the regional background.

Statistical methods

Regarding statistical measures of agreement between data sets, Robinson (1957) criticized the use of the coefficient of determination (r^2) in comparisons of observed an theoretically deduced values of a variable for the purpose of model validation, as it measures the degree to which paired values of two variables X_1 and

X_2 are proportional ($X_1 = a + bX_2$) rather than identical ($X_1 = X_2$) to each other. Later, Willmott (1981) and Willmott *et al*. (1985) criticised what already at the time was described as "traditional methods of evaluating geographical models by statistical comparisons between observed and simulated variates, particularly the coefficient of determination (r^2)". There are two underlying assumptions regarding the significance test associated with the coefficient of determination. First, both variables must be normally distributed, and second, the data must be a random sample.

The observed and predicted daily averaged time series data we used were not normally distributed (Kahn 1973, also verified at http://contchart.com/goodness-of-fit.aspx), but more closely resemble the log-normal distribution. On the other hand, our aim was to study the AQ modelling system predicted data agreement with the observed data, not to build a regression model.

Instead of using r^2, Willmott *et al*. (1981) suggested the use of a root mean squared error (RMSE) and an index of agreement (d). They first defined the index of agreement as

$$d_2 = 1 - \frac{\sum_{i=1}^{n}\left(P_i - O_i\right)^2}{\sum_{i=1}^{n}\left(\left|P_i - \overline{O}\right| + \left|O_i - \overline{O}\right|\right)^2}, \qquad (1)$$

where n is the sample size, P is the predicted quantity, P_i is the value of P at time i, O is the observed quantity, O_i is the value of O at time i, and $\overline{O}$ is the mean value of the time series O_i (i = 1, ..., n). d_2 is dimensionless and has the range of 0–1.

The more well-known RMSE is defined as

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(P_i - O_i\right)^2}. \qquad (2)$$

The RMSE has the same metric as the observed and predicted quantities.

Later, Willmott *et al*. (1985, 2011) decided to develop the concept further, as they felt that the squaring of errors in Eq. 1 over-weighted the influence of those errors. Therefore, they introduced the modified index of agreement, defined as

$$d_1 = 1 - \frac{\sum_{i=1}^{n}\left|P_i - O_i\right|}{\sum_{i=1}^{n}\left(\left|P_i - \overline{O}\right| + \left|O_i - \overline{O}\right|\right)}, \qquad (3)$$

where d_1 is dimensionless and has the range of 0–1.

Index of agreement-type statistics are not measures of correlation or association in the formal sense. Instead, they provide a measure of the degree to which the model predictions are error free. We studied the behaviour of d_1 with the data set containing the natural logarithms of six of our observed daily-averaged time series *versus* random time series (with the same means and standard deviations as the observed time series). In this comparison d_1 was between 0.28 and 0.32. Regarding d_2, Kukkonen *et al*. (2000b) stated that "Numerical experiments have shown that a totally random predicted time series having the same range of variability as the measured time series, would result in an index of agreement value equal to approximately 0.4." Variability means dispersion of the data, a common measure of which is the standard deviation. Both d_1 and d_2 approach the value of 1 when the agreement of observed and predicted time series increases, and d_1 approaches 1 slower than d_2. Compared with the coefficient of determination, d_1 and d_2 are more strict and especially sensitive to the difference of the means of the observed and predicted data sets. The d_2 index of agreement has often been used in recent air quality modelling studies but was denoted there as IA (e.g. Kauhaniemi *et al*. 2011 and 2014, Singh *et al*. 2014). Here, we calculated d_2 and d_1 to facilitate comparisons.

RMSE is seen an a useful measure of the model error to be used with d_2, but Willmott *et al*. (2011) introduced two more error statistics to be used with d_1: the mean-absolute error MAE and mean-absolute deviation, defined as

$$\mathrm{MAE} = \frac{1}{n}\sum_{i=1}^{n}\left|P_i - O_i\right|, \qquad (4)$$

$$\mathrm{MAD} = \frac{1}{n}\sum_{i=1}^{n}\left|O_i - \overline{O}\right|. \qquad (5)$$

The other statistics we used are a Fractional Bias, given by

$$\mathrm{FB} = \frac{2\left(\overline{P} - \overline{O}\right)}{\overline{P} + \overline{O}}, \qquad (6)$$

where $\overline{P}$ is the mean value of the time series P_i (i = 1, ..., n), and the Fraction of Two, defined as

F2 = fraction of data for which $0.5 \le P/O \le 2$. (7)

Results and discussion

Comparison of observed and predicted concentrations

We first compared the regional-background predictions against the nearest available regional-background measurements. The predictions of the LOTOS-EUROS model were lower by 12% and 22% than the annual Luukki regional background concentration averages in 2008 for $PM_{2.5}$ and PM_{10}, respectively. These percentages were based on the measured annual average values, the $PM_{2.5}$ value at the regional background station of Luukki (7.13 μg m^{-3}) and the PM_{10} value at the regional background station of Virolahti (9.13 μg m^{-3}). The reasons for this under-prediction are discussed in more detail in Kukkonen *et al*. (2016). Briefly, the omission of biogenic secondary aerosol contributions and the underestimation of PM from natural sources have been recognized. The performance of the LOTOS-EUROS model in international inter-comparisons has been comparable with other European chemical-transport models (Stern *et al*. 2008, Solazzo *et al*. 2012).

There were five stations representing urban environment in the HMA where measurements of PM_{10} were carried out in 2008. The station in Kallio is an urban background site (Fig. 2a), and the other of the stations (Fig. 2b–e) are urban traffic sites. The temporal variation of the predictions agrees fairly well with that of the observations; however, most of the predicted values were underestimations. However, the highest values in spring, during the most intensive road dust season, were in most cases reasonably well predicted by the model.

Monitoring measurements of $PM_{2.5}$ were available from two urban environment stations in 2008 (Fig. 3) and five such stations in 2010 (Fig. 4).

Comparisons of the measured *versus* predicted daily concentrations (Table 2) indicated that the agreement of the measured and predicted hourly time-series can be considered fairly good for the modelling system including the LOTOS-EUROS model, and good when using measured regional-background concentration values from the available measurement sites. For instance, d_1 for 2008 varied from 0.57 to 0.57 and from 0.47 to 0.51, for $PM_{2.5}$ and for PM_{10}, respectively. However, only two measurement stations were available for $PM_{2.5}$ in 2008; the d_1 range for $PM_{2.5}$ mentioned above may therefore not be representative for the predictions covering the whole of the metropolitan area. The d_1 and F2 values (for $PM_{2.5}$ at Kallio and Mannerheimintie) were higher for 2010 than for 2008. A better temporal agreement in 2010 was mainly caused by the use of measured regional-background values, instead of the regional scale model predictions.

The $PM_{2.5}$ and PM_{10} concentrations in 2008 were under-predicted, as indicated by the negative FB values. This was caused mainly by the under-predicted regional-background concentrations. There was no systematic under- or over-prediction of the $PM_{2.5}$ values in 2010. The under-prediction in 2008 was more pronounced for PM_{10} than for $PM_{2.5}$. The greater under-prediction of PM_{10} was caused by the under-predicted regional background, and the uncertainties in the modelling of suspension of road dust.

The agreement of predictions and measurements can also be examined in terms of the various stations and the categories of stations. The temporal agreement of predictions and data is better for the urban background station (Kallio), than for the urban traffic stations. This could be caused by various factors at the urban traffic stations. First, the dispersion modelling system did not allow for the effects of buildings and other obstacles. Second, there were also several other specific features for one or more of the urban traffic stations that could not be explicitly modelled. In particular, at the urban traffic site of Mannerheimintie, street reconstruction works were in progress. Also the streets at this site are paved with cobblestones the effect of which on the emissions of suspended dust could not be evaluated.

In a previous comparison of the observed PM_{10} concentrations in 2004 (116 data points from one location) and the predictions of the street canyon model OSPM combined with the FORE model daily averaged data from a street canyon (Kauhaniemi *et al*. 2011), based on 116 data points from one location, d_2 = 0.87, F2 = 0.94 and FB = 0.03. In our comparison of the observations in the year 2008 (364–366 data points from five urban locations) with the modelling system including the road traffic dispersion

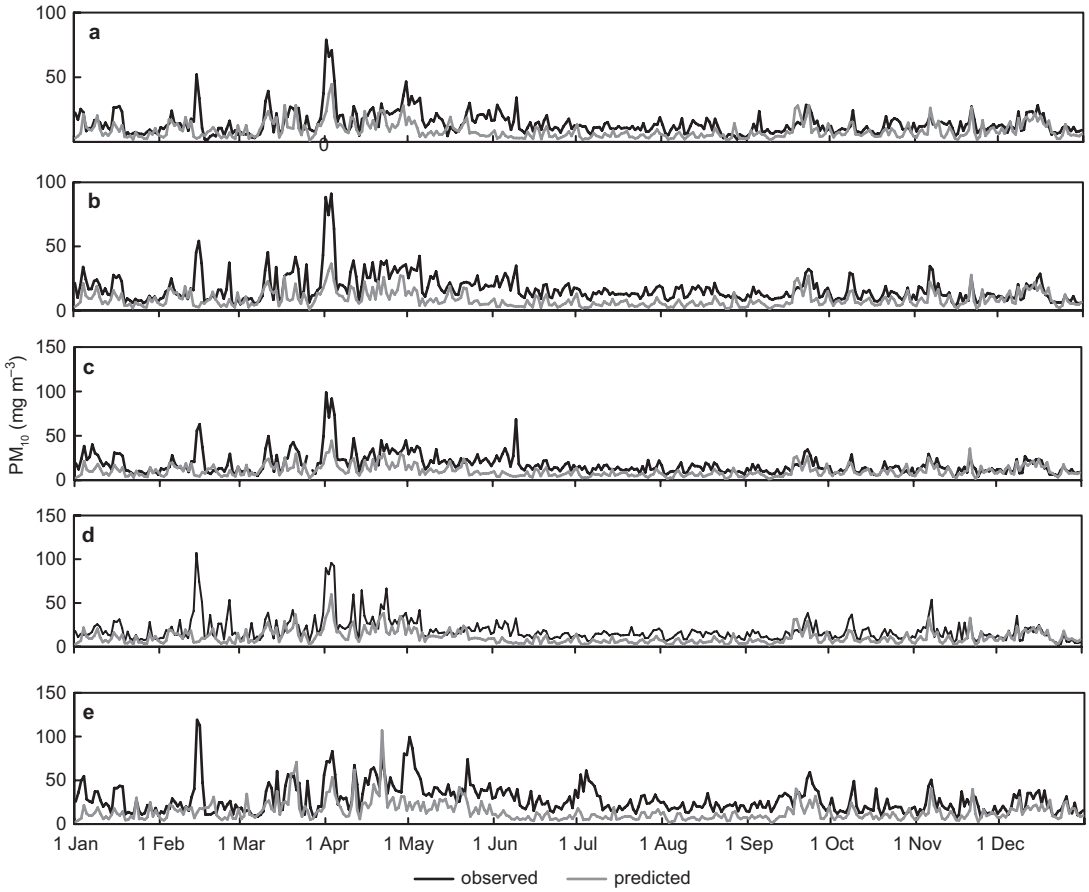

Fig. 2. Daily-averaged observed and predicted time series of PM_{10} at five sites in 2008. Measurement site categories: arban background (**a**) Kallio, and urban traffic: (**b**) Mannerheimintie, (**c**) Vallila, (**d**) Leppävaara and (**e**) Tikkurila.

model and the FORE model, d_2 for PM_{10} varied between 0.60 and 0.66.

The measured concentration values (two-week averages, 57 data points) of $PM_{2.5}$ within the ESCAPE project were also compared with the predictions (Fig. 5) for the 13 ESCAPE sites in the HMA, which yielded $d_1 = 0.79$, FB = 0.1 and F2 = 1.

Source contributions to the $PM_{2.5}$ and PM_{10} concentrations

The predicted source contributions of long range transport (LRT), vehicular traffic exhaust and road dust suspension on $PM_{2.5}$ and PM_{10} concentrations in 2008 were considered (Table 3). The vehicular cold start and cold driving emissions were also included in the category of vehicular exhausts. For convenience, average values computed based on values at four urban traffic stations are presented.

The LRT was responsible for most of the pollution in both PM fractions at urban-background and urban-traffic stations. For instance, the LRT contribution at the urban traffic stations was on the average 81% (range from 72% to 92%) and 67% (range from 50% to 83%) for $PM_{2.5}$ and PM_{10}, respectively. As expected, those values were even higher for the urban-background station, 92% and 82%, and the LRT contributions were greater in case of $PM_{2.5}$ than PM_{10}.

The contributions of vehicular traffic exhaust emissions ranged from 5% to 20%, and from 4% to 16%, for $PM_{2.5}$ and PM_{10}, respectively. The vehicular suspension contributions ranged

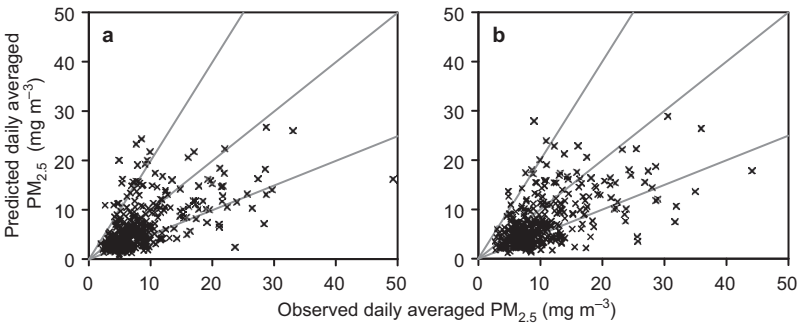

Fig. 3. Scatter plots of the daily averaged measured and predicted concentrations of $PM_{2.5}$ in 2008 at (**a**) Kallio and (**b**) Mannerheimintie.

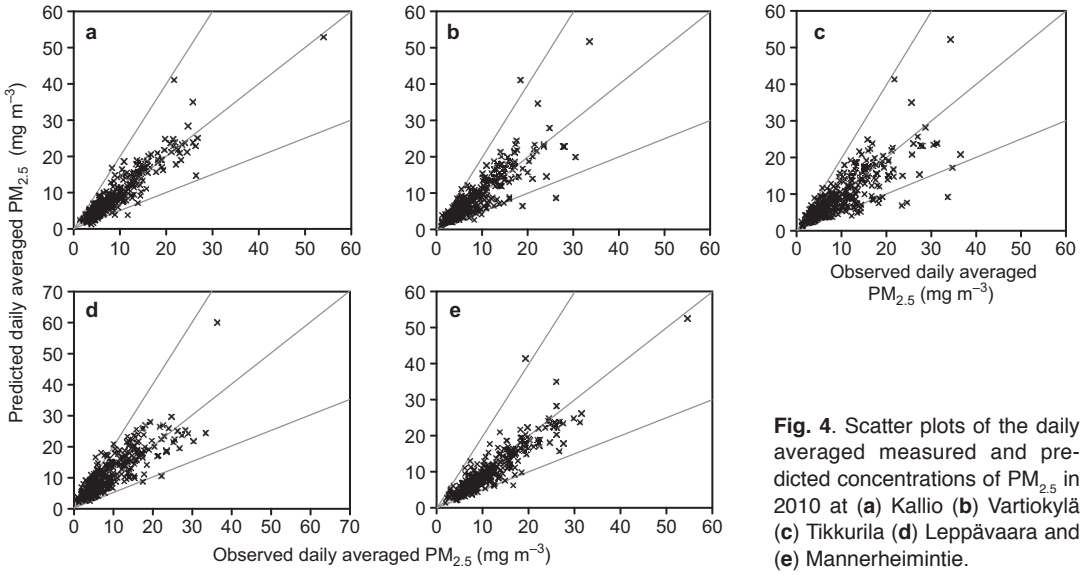

Fig. 4. Scatter plots of the daily averaged measured and predicted concentrations of $PM_{2.5}$ in 2010 at (**a**) Kallio (**b**) Vartiokylä (**c**) Tikkurila (**d**) Leppävaara and (**e**) Mannerheimintie.

from 2% to 8%, and from 12% to 38%, for $PM_{2.5}$ and PM_{10}, respectively. The contribution from suspension emissions was greater than that of the exhaust emissions in case of PM_{10}, and *vice versa* for $PM_{2.5}$. At the urban traffic sites, vehicular traffic was on average (including both exhaust and suspension emissions) responsible for 20% and 33% of the concentrations of $PM_{2.5}$ and PM_{10}, respectively.

The source contributions of $PM_{2.5}$ in the HMA wer previously studied using a more limited data set (from two sites during weekdays in 2002) by Kauhaniemi *et al*. (2008). They evaluated the LRT contribution using a semi-empirical method of Kukkonen *et al*. (2008). They found the contributions of LRT, vehicular exhaust and vehicular suspension emissions at the site of Vallila to be 61%, 13% and 23%, respectively, while in this study for 2008 they were 77%, 20% and 3%, respectively. However, the percentage contributions at one specific station can vary substantially from year to year, depending on the meteorological conditions and in case of suspension emissions, also on numerous other factors, such as, e.g., traction control and street cleaning methods.

Those values can qualitatively be compared also with the corresponding results from other Nordic countries. Gidhagen *et al*. (2013) reported that the modelled residential PM_1 and PM_{10} were dominated by LRT, whereas urban sources contributed about 30% of the PM_{10} concentrations in Stockholm. Laupsa *et al*. (2009) reported the source contributions to $PM_{2.5}$ for a roadside site in Oslo in winter of 2004. The values obtained by using dispersion and receptor modelling were 18%–28% for LRT, 10%–14%

Table 2. Statistical analysis for the comparisons of predicted (C_{pred}) *versus* observed (C_{obs}) daily averaged concentrations (μg m^{-3}). The number of the daily averaged values included (*n*) and the fraction of the hourly measurement data available for the calculations of the daily averages (*f*) are also presented. Values given for C_{obs} and C_{pred} are means ± SDs.

Year	Pollutant	Statistical measure	Station (*category)						
			Luukki (RB)	Kallio (UB)	Mannerheimintie (UT)	Leppävaara (UT)	Tikkurila (UT)	Vallila (UT)	Vartiokylä (ResB)
2008	$PM_{2.5}$	d_1	0.56	0.57	0.53				
		d_2	0.73	0.72	0.69				
		FB	–0.12	–0.23	–0.32				
		F2	0.72	0.69	0.70				
		n	363	361	365				
		f	0.94	0.96	0.94				
		C_{obs}	7.1 ± 5.13	8.57 ± 5.90	10.44 ± 6.08				
		C_{pred}	6.28 ± 4.63	6.79 ± 4.94	7.54 ± 5.05				
		MAE	3.24	3.72	4.47				
		MAD	3.39	4.03	4.31				
		RMSE	4.7	5.36	6.03				
	PM_{10}	d_1		0.5	0.47	0.51	0.47	0.51	
		d_2		0.66	0.60	0.63	0.60	0.62	
		FB		–0.48	–0.65	–0.53	–0.48	–0.53	
		F2		0.57	0.45	0.57	0.48	0.61	
		n		364	366	364	366	365	
		f		0.96	0.99	0.98	0.99	0.98	
		C_{obs}		14.28 ± 9.31	28.15 ± 16.57	18.65 ± 13.43	16.9 ± 10.87	17.87 ± 11.94	
		C_{pred}		8.68 ± 6.26	14.32 ± 11.23	10.81 ± 7.68	8.59 ± 5.90	10.42 ± 6.45	
		MAE		6.90	15.07	8.92	8.78	8.40	
		MAD		6.38	12.4	8.45	7.39	8.12	
		RMSE		9.47	19.88	13.55	12.00	12.29	
2010	$PM_{2.5}$	d_1	0.998	0.82	0.80	0.69	0.74		0.78
		d_2	1	0.96	0.95	0.90	0.89		0.92
		FB	0.001	0.01	–0.12	0.19	–0.04		0.05
		F2	1	0.99	0.98	0.89	0.93		0.96
		n	360	360	360	350	360		357
		f	0.91	0.90	0.88	0.87	0.91		0.88
		C_{obs}	8.52 ± 6.27	9.04 ± 5.80	11.08 ± 6.36	8.87 ± 6.24	9.54 ± 6.67		8.25 ± 5.62
		C_{pred}	8.53 ± 6.27	9.13 ± 6.38	9.84 ± 6.30	10.78 ± 6.59	9.07 ± 6.33		8.58 ± 6.25
		MAE	0.02	1.59	2.06	3.05	2.63		2.02
		MAD	4.63	4.29	4.79	4.83	5.18		4.42
		RMSE	0.04	2.41	2.92	4.01	4.13		3.20

* RB = region background, UB = urban background, UT = urban traffic, ResB = residential background.

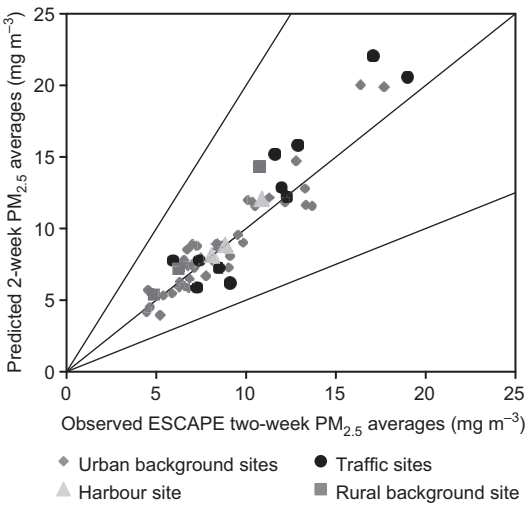

Fig. 5. Comparison of the two-week averages of measured (in the ESCAPE project) and modelled concentrations of $PM_{2.5}$ in 2010.

for soil, salt and road dust, and 6%–24% for road vehicle exhaust.

Concerning contributions of omitted sources, major stationary sources (energy production and industry) in the HMA were previously evaluated to affect concentrations by only 1%–2% at the urban scale (Soares *et al*. 2014). It has previously been shown with STEAM2 shipping emission modelling (Jalkanen *et al*. 2012, Johansson *et al*. 2013) that the contribution of primary shipping emissions to the total concentrations of $PM_{2.5}$ in the HMA is on average about 3%. However, this contribution can be higher than 20% in the vicinity of the harbours (within a distance of approx. 1 km).

Based on measurements and source apportionment, Saarnio *et al*. (2012) evaluated the contribution of local-scale wood burning to $PM_{2.5}$ concentrations to be notable at some locations in the HMA during the cold season (October–March). They estimated that the small-scale combustion was responsible for 18%–29% and 31%–66% of the $PM_{2.5}$ concentrations during the cold season at three urban-background and two suburban sites, respectively. Residential wood burning emission has spatially heterogeneously distributed area-like sources in the HMA. This makes their concentration and source contribution highly location-dependent.

Table 3. Predicted annual average total concentrations and source contributions of the long range transport (LRT) vehicular exhaust and suspended $PM_{2.5}$ and PM_{10} at the available measurement stations in 2008.

PM species	Source	Station (category*)						
		Luukki (RB)	Kallio (UB)	Average of UT stations	Mannerheimintie (UT)	Leppävaara (UT)	Tikkurila (UT)	Vallila (UT)
$PM_{2.5}$	Total (μg m^{-3})	6.28	6.8	7.8	7.5	8.7	6.8	8.1
	LRT (%)	99.84	92	81	83	72	92	77
	Exhaust (%)	0.1	5	15	12	20	6	20
	Suspension (%)	0.06	2	5	5	8	2	3
PM_{10}	Total (μg m^{-3})	7.17	8.7	11.0	10.8	14.3	8.6	10.4
	LRT (%)	99.5	82	67	66	50	83	69
	Exhaust (%)	0.09	4	10	8	12	5	16
	Suspension (%)	0.42	14	23	25	38	12	16

* RB = region background, UB = urban background, UT = urban traffic.

Road traffic suspension emissions have a strong monthly variation, while emissions from tyre and brake wear are probably temporally more uniform.

The spatial distributions of the $PM_{2.5}$ and PM_{10} concentrations

The predicted annual average PM concentrations in 2008 ranged from 6.4 to 14.4 μg m^{-3} for $PM_{2.5}$ and from 7.4 to 33.9 μg m^{-3} PM_{10}. The major traffic networks, including two ring roads, the main highways and the centre of Helsinki, where the highest concentrations occurred were clearly distinguishable (Fig. 6a–b). Although the roads and streets are continuous lines, there are several possible reasons for "patchiness" of the concentration sources. Traffic maxima at junctions produce local concentration maxima also along the line sources. Meteorology, especially the variation of wind direction, also affects the dispersion of the emissions and may produce local maxima. Finally, concentrations were modelled for a point grid and the concentrations between the points were produced by interpolation.

Summary and conclusions

The suspended road dust has been see as an important source category especially in northern and mountainous regions. However, with an increasing vehicular traffic intensities and continuously decreasing exhaust emissions per vehicle, suspended road dust has become important in practically all road traffic environments. However, due to their complexity, detailed road suspension emission models have previously been applied only to specific street segments, instead of extensive urban areas (e.g., Kauhaniemi *et al.* 2011, 2014). We included the road dust suspension model FORE (Kauhaniemi *et al.* 2011) to an urban modelling system, and applied that system to the whole HMA. This procedure provided quantitative information on the contributions of suspended dust to urban PM mass-based concentrations.

In this study, the regional-background concentrations were either evaluated using the chemical transport model LOTOS-EUROS (for the year 2008), or by using measured regional-background concentrations (for the year 2010). The modelled results were compared with the measurement data of the Helsinki Region Environmental Services Authority and those measured in the ESCAPE (European Study of Cohorts for Air Pollution Effects) project. The modelled long-range-transported contribution to PM was also compared with observations.

According to d_1, the agreement of the measured and predicted daily time-series can be considered fairly good for the modelling system including the LOTOS-EUROS regional-background model. The $PM_{2.5}$ concentrations were slightly and the PM_{10} concentrations substantially under-predicted for 2008, which was caused mainly by the under-predicted regional-background concentrations and the known under-prediction of the suspended dust by the model FORE (Kauhaniemi *et al.* 2014). The greater under-prediction for PM_{10} as compared with that for $PM_{2.5}$ may also have been partly caused by uncertainties in the modelling of the suspension of road dust for PM_{10}. The predicted and measured two-week averages at ESCAPE sites agreed well. However, due to the major contributions of the LRT, this comparison of modelled and measured results was not a critical test of the urban-scale modelling system.

The contributions of LRT and vehicular traffic to the measured concentrations had not earlier been evaluated and analysed for this urban area. The contribution of LRT was the main factor for both PM fractions, both at urban-background and urban-traffic stations. The vehicular-suspension contributions at all the available urban-background and urban-traffic stations ranged from 2% to 8% and from 12% to 38%, for $PM_{2.5}$ and PM_{10}, respectively. The contribution originating from suspension emissions was greater than that of the exhaust emissions in case of PM_{10}, and *vice versa* for $PM_{2.5}$.

Clearly, there are several uncertainties that affect the agreement of measured and modelled values. The dispersion modelling system as applied in this study did not explicitly allow for the effects of buildings and other obstacles on the atmospheric dispersion. We also did not include in the dispersion calculations the contri-

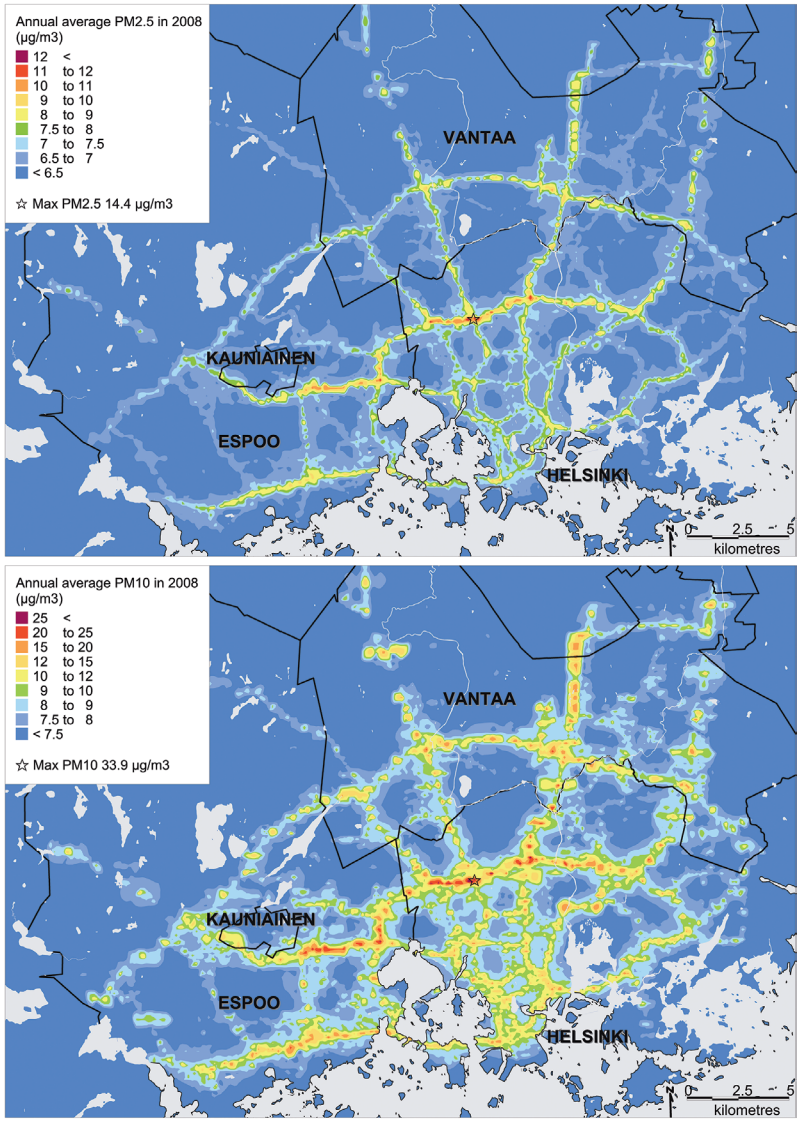

Fig. 6. The predicted spatial distributions of the annual average concentrations (μg m^{-3}) of $PM_{2.5}$ (top) and PM_{10} (bottom) in the Helsinki Metropolitan area in 2008.

butions from residential wood combustion, shipping, major stationary sources, or tyre, brake, and clutch wear. However, based on previous studies and available data, we evaluated the significance of their contribution to both emissions and concentrations.

The measurement stations used in our study here were not located in street canyons, i.e., urban locations with a substantial ratio of building height to street width. The site that resembles a street canyon the most was the urban-traffic site at Mannerheimintie where the height of the surrounding buildings is approximately 20 m and the street width is 47 m resulting in an aforementioned ratio of < 0.5.

Some of the included stations had special characteristics that we could not model, e.g., street reconstructions in the vicinity of the urban traffic-site in Mannerheimintie.

In ongoing and future studies, we are focusing and will focus especially (i) on analysing in more detail the emissions and dispersion from small-scale combustion, shipping and harbours, (ii) on analysing dispersion in street canyons, especially of suspended dust and other non-exhaust particulate matter, (iii) on analysing fine

particulate matter, particle number concentrations and the chemical composition of particles, and (iii) on modelling the regional background concentrations more accurately, including also the effects of secondary organic matter and various non-anthropogenic sources.

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